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1,3,5-Trialkyl-hexahydro-1,3,5-triazines–*N*-methylenealkylamines equilibria. ^{1}H NMR studies in solutions

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ABSTRACT

The behavior of 1,3,5-trialkyl-1,3,5-hexahydrotriazines (**A**) in a variety of solvents was investigated by ^{1}H NMR. **A** are stable for linear alkyls in deuterated solvents such as chloroform, benzene, acetone, dioxane, dimethylsulfoxide and acetonitrile. In case of branched alkyls, **A** are in equilibrium with *N*-methylenealkylamines (**B**). The **A**/**B** ratio depends on solvent, concentration of the sample and temperature. **A** react easily with methanol and lead to the formation of *N*-(methoxymethyl)amines (**C**), which are in equilibrium with **B**. Both the quantitative evaluation of **A**–**B** equilibrium in solutions as well as the formation of **C** in methanol was described for the first time.

1. Introduction

1,3,5-Trialkyl-hexahydro-1,3,5-triazines (**A**) (Scheme 1) are reported to be the stable products of primary aliphatic amine and formaldehyde condensation [1–7]. The methods of their synthesis have been extensively reviewed in 1959 by Smolin and Rapoport [1]. **A** are reported to be stable in neutral or alkaline solutions [8], but in the presence of aqueous acids readily decompose with the formation of formaldehyde and the primary amine salt [4,5]. Under the action of HCl in anhydrous media they form imminium salts [9]. Pyrolysis of **A** results in the formation of *N*-methylenealkylamines (**B**), stable at low temperature [10–12]. Some of **B** were spectroscopically characterized at −60 °C [13]. At higher temperatures they spontaneously trimerize. Due to their cyclic structure, 1,3,5-trialkyl-hexahydro-1,3,5-triazines display temperature-dependent ^{1}H NMR spectra in the range of ring methylene signals [14]. When heated with sodium methoxide in methanol solution **A** give *N*-(methoxymethyl)amines (**C**). It is also possible to perform the reverse reaction by heating **C** under reduced pressure [15] (Scheme 1).

The **A**–**B** equilibrium in solution has been deduced as early as in 1932 [4] and later on taken as a fact [16–18]. Nevertheless, no ^{1}H NMR spectra with signals corresponding to both **A** and **B** forms have been described. The equilibrium was investigated by means of IR in various solvents [3]. The monomeric form was reported to be more stable in acidic media, but no quantitative assessment has ever been done. 1,3,5-Trialkyl-hexahydro-1,3,5-triazines form complexes with metal ions [19–22], some of the complexes have been applied as polymerization catalysts [20–22]. The most important synthetic application of **A** is their use as precursors of *N*-methylenealkylamines (**B**) [9,18,23–32]. As such, **A** are applied in the formal context of Mannich reaction [9,15,32] or other aminomethylation reactions [2,15]. The structures and concentrations of the species in solutions should be crucial in their synthetic applications, so we found it interesting to investigate the behavior of **A** in various solvents.

2. Experimental

^{1}H NMR spectra were taken on Varian Gemini 200 MHz or Varian Unity Plus 200 MHz (0 relaxation delay). IR spectra were recorded on Perkin Elmer PE-577. Elemental analyses were carried out on Perkin Elmer CHNS/O II Model 240. Several 1,3,5-trialkyl-hexahydro-1,3,5-triazines were obtained by a known method of formaldehyde and primary amine condensation [1,2,10,32] and investigated as crude oily product. The IR spectra of the crude 1,3,5-trialkyl-hexahydro-1,3,5-triazines in condensed phase (thin film or KBr pellet) revealed no bands in the range of 1685–1580 cm^{-1} corresponding to C=N bond in the monomeric structure. A crystalline 1,3,5-tricyclohexyl-hexahydro-1,3,5-triazine (**TCH**) and its liquid linear alkyl isomer (**TH**), purified by crystallization from acetone (**TCH**) and vacuum distillation (**TH**) were investigated as model compounds. The purity of the compounds was confirmed on the basis of elemental analysis (calculated for $C_{21}H_{39}N_3$ (**TCH** and **TH**) C: 75.62, N: 12.62, H: 11.76, measured

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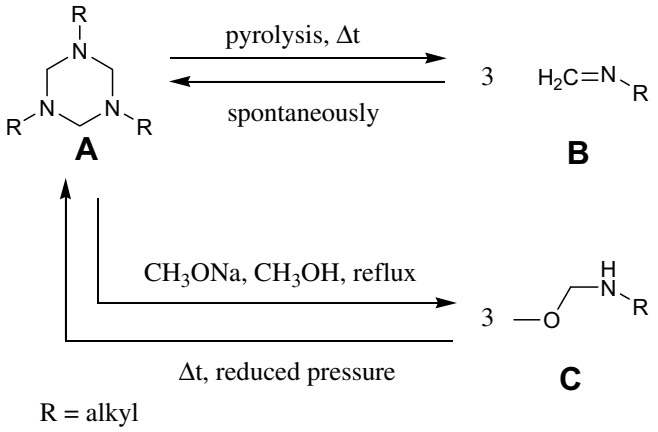


Scheme 1.

C: 75.59, N: 12.55, H: 11.62 (**TCH**) and C: 75.10, N: 12.89, H: 12.00 (**TH**) as well as IR spectra. Solutions of the compounds were prepared in a variety of deuterated solvents and kept at room temperature for 1 h.

3. Results and discussion

The behavior of 1,3,5-trialkyl-hexahydro-1,3,5-triazines in chloroform solutions is dependent on the alkyl substituents. For branched alkyls such as *t*-Bu, *i*-Pr and cyclohexyl a mixture of **A** and **B** was observed, whereas in case of linear alkyls such as *n*-Bu, *n*-Pr and *n*-hexyl no signals characteristic of monomeric forms were present. Moreover, considerable differences in reactivity of linear and branched 1,3,5-trialkyl-hexahydro-1,3,5-triazines and influence of the solvent and temperature applied could be observed in the Mannich reaction of phenols with 1,3,5-tricyclohexyl-hexahydro-1,3,5-triazines [32]. A crystalline 1,3,5-tricyclohexyl-hexahydro-1,3,5-triazine (**TCH**) was chosen as a model substance due to its well defined structure, that was previously determined by X-ray [10]. The linear alkyl isomer (**TH**) was investigated for comparison.

The characteristic signals corresponding to the methylene groups of heterocyclic ring (*a*) in **TH** and **TCH**, the CH_2=N group (*b*) in *N*-methylenealkylamines **B** and O–CH_2–N (*c*) in *N*-(methoxymethyl)amines **C** were observed in solutions (Fig. 1 and Table 1). The equilibrium concentrations of the species were calculated on the basis of the dissolved amount of **A** and the contents of the species in solutions evaluated by means of integration of the corresponding signals without internal standard.

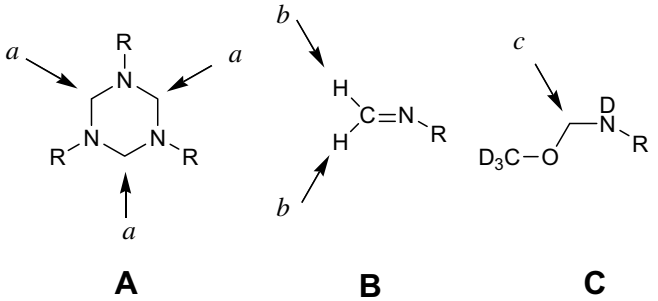


Fig. 1. Characteristic groups of **A**, **B** and **C** species.

Table 1
^{1}H chemical shifts observed in solutions of **TH** and **TCH** in various solvents at room temperature

Entry	Compound	Solvent	δ_a (ppm)	δ_b (ppm)	δ_c (ppm)
1	**TCH**	$CDCl_3$	3.60	7.45, 7.05	–
2	**TCH**	$(CD_3)_2CO$	3.53	7.43, 6.99	–
3	**TCH**[b]	$(CD_3)_2SO$	3.32	7.43, 7.02	–
4	**TCH**[b]	CD_3CN	3.49	7.42, 7.00	–
5	**TCH**	C_6D_6	3.66	6.99, 6.82[a]	–
6	**TCH**	$C_4D_8O_2$	3.39[a]	7.26, 6.86	–
7	**TCH**	CD_3OD	4.17	7.45, 7.08	4.32
8	**TH**	$CDCl_3$	3.28	–	–
9	**TH**	$(CD_3)_2CO$	3.27	–	–
10	**TH**	$(CD_3)_2SO$	3.16	–	–
11	**TH**	CD_3CN	3.36	–	–
12	**TH**	C_6D_6	3.40	–	–
13	**TH**	$C_4D_8O_2$	3.23	–	–
14	**TH**	CD_3OD	4.14	–	4.26

[a] Partly overlapped with a solvents signal.
[b] Low solubility of **TCH**.

3.1. The influence of the sample concentration on the *A–B* equilibrium in inert solvents

The behavior of 1,3,5-trialkyl-hexahydro-1,3,5-triazines in inert solvents depends on the alkyl substituent. In the case of linear alkyl group, **A** are very stable and make the only form observed at the investigated concentrations and temperatures. In the case of the cyclohexyl substituents, a mixture of **A** and **B** was observed in all the applied inert solvents. ^{13}C NMR spectra of **TH** in $CDCl_3$ also revealed exclusively signals corresponding to **A**, whereas in case of **TCH**, a signals of **B** (including the most characteristic one at 150 ppm corresponding to C=N group) could also be observed.

Due to good solubility of the investigated compounds and no coincidence with solvent's signals, chloroform and acetone were chosen as solvents appropriate for further studies. Samples of various $[A_0]$ in acetone and chloroform were prepared. The relative A/B molar ratio was estimated by integration of the corresponding signals without internal standard. It was assumed that the **A–B** equilibrium is the only process taking place in the sample and $a + b = 1$, where a and b are molar fractions of **A** and **B** respectively. The equilibrium molar concentrations of **A** and **B** were calculated on the basis of the following Eqs. (1) and (2).

$$[A] = 3 \times a \times [A_0]/(3 \times a + b) \quad (1)$$

$$[B] = 3 \times b \times [A_0]/(3 \times a + b) \quad (2)$$

On the basis of [A] and [B], the equilibrium constants have been calculated following Eq. (3).

$$K_{A,B} = [B]^3/[A] \quad (3)$$

The calculated amounts of **A** and **B** depend on the solvent and concentration of the sample (Table 2 and Fig. 2).

Table 2
Initial concentration of **A**, equilibrium concentration of **A** and **B**, equilibrium constants in $(CD_3)_2CO$ and $CDCl_3$ solutions

Entry	$(CD_3)_2CO$				$CDCl_3$		
	$[A_0]$ $[\times 10^4/(\text{mol dm}^{-3})]$	[A]	[B]	$K_{A,B} \times 10^9$	[A] $[\times 10^4/(\text{mol dm}^{-3})]$	[B]	$K_{A,B} \times 10^9$
1	4.4	3.9	1.7	12.6	3.5	2.6	50.2
2	2.8	2.4	1.3	9.2	2.1	2.1	44.1
3	2.0	1.6	1.1	8.3	1.5	1.6	27.3
4	1.2	0.89	0.93	9.0	0.75	1.3	29.3
5	0.4	0.22	0.55	7.6	0.19	0.63	13.2[a]
6	0.2	0.08	0.37	6.3	0.03	0.52	46.9

[a] The result was reproducibly obtained in several independent experiments.

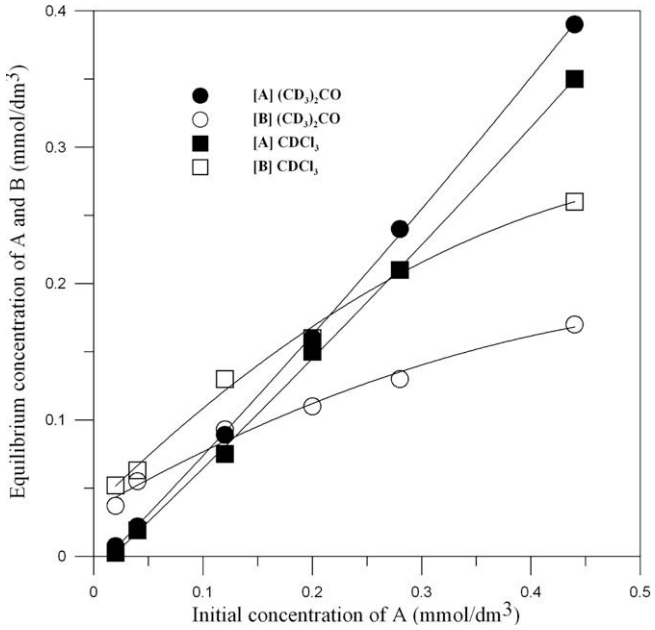


Fig. 2. Equilibrium concentrations of **A** and **B** in $(CD_3)_2CO$ and $CDCl_3$ solutions depending on the initial concentration of **A**.

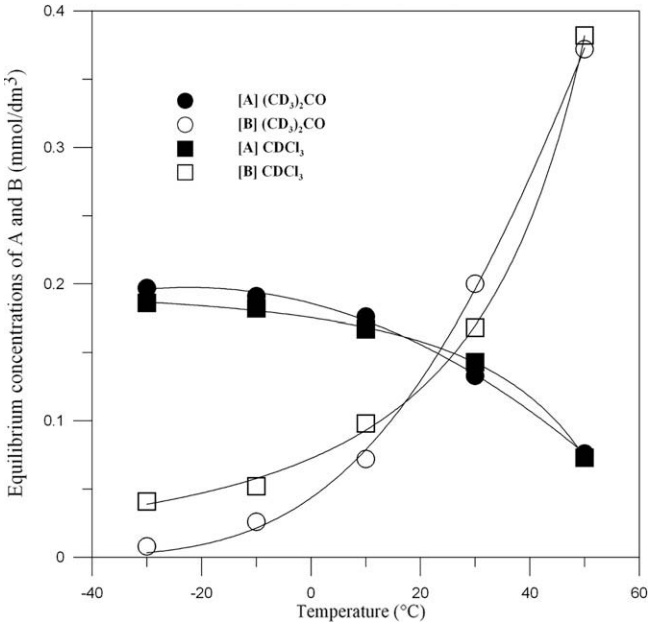


Fig. 3. Equilibrium concentrations of **A** and **B** depending on temperature.

A similar tendency in the changes of the equilibrium concentrations of **A** and **B** in $(CD_3)_2CO$ and $CDCl_3$ solutions was observed. At low [A_0], **B** is the predominant form in solution, whereas at higher [A_0] **A** is in majority. The observation explains some contradictory reports on the spectral properties of 1,3,5-trialkyl-hexahydro-1,3,5-triazines. For example Jewett et al. [33] report no **B** form in a variety of investigated solutions. To the contrary, Müller et al. [34] gave exclusively shifts corresponding to **B** to characterize 1,3,5-trialkyl-hexahydro-1,3,5-triazines by ^{1}H NMR. Similarly, the reported rapid decomposition of the 1,3,5-tri-*tert*-butyl-hexahydro-1,3,5-triazine in $CDCl_3$ solution [14] might in fact be the result of low concentration of the sample.

On the basis of the changes of the concentration of **A** it was possible to evaluate the average equilibrium constants of the observed transformation which is about 9×10^{-9} in deuterated acetone and about 4×10^{-8} in $CDCl_3$. The shift of the equilibrium in $CDCl_3$ can be caused by acidic impurities resulting from partial decomposition of the solvent.

3.2. The influence of temperature on the *A*–*B* equilibrium in inert solvents

The influence of the temperature on equilibrium concentrations of **A** and **B** was also investigated. Solutions of **TCH** in $CDCl_3$ and $(CD_3)_2CO$ were prepared (2×10^{-4} mol dm^{-3}). The spectra were taken at temperatures ranging from −30 to +50 °C. The samples were kept at the desired temperature and monitored by ^{1}H NMR every 15 min, until no considerable changes in their compositions could be detected. The equilibrium concentrations of **A** and **B** (Table 3 and Fig. 3) were evaluated according to the previously described methodology.

Changes of the equilibrium concentrations depending on temperature are very similar for acetone and chloroform. At +10 °C and lower temperatures, **A** was the predominant form in both of the solvents. At +30 °C and higher temperatures **B** was in majority. The observed changes of the spectra with the temperature confirm the existence of the expected cyclic structure in solution. At −10 °C, broadening of signal corresponding to methylene linkages (3.60 ppm), due to the slowing of the chair to-chair interconversion of the cyclohexane backbone, was observed in both of the investigated solvents. At −30 °C two broadened signals were observed instead of one. The rate constant of the process at the coalescence temperature (−15 ± 0.5 °C in $CDCl_3$) was evaluated according to the following Eq. (4).

$$k_e = (3.14 \times 164\ \text{Hz})/1.41 = 365\ \text{s}^{-1} \quad (4)$$

The value of free energy of activation at the coalescence temperature (50.35 kJ/mol = 12.02 kcal/mol), calculated following the Eyring equation is in agreement with the ones reported for analogous compounds [33]. In the spectrum taken in $CDCl_3$ at −50 °C, two doublets (at 4.04 and 3.22 ppm) corresponding to equatorial and axial hydrogens were present (Fig. 4).

Table 3
Calculated concentrations of **A** and **B** depending on temperature

Entry	Temperature	$(CD_3)_2CO$			$CDCl_3$		
		[A] [$\times 10^4$/(mol dm^{-3})]	[B]	$K_{A,B}$	[A] [$\times 10^4$/(mol dm^{-3})]	[B]	$K_{A,B}$
1	−30	1.97	0.08	2.60×10^{-12}	1.86	0.40	3.44×10^{-10}
2	−10	1.91	0.26	9.20×10^{-11}	1.82	0.52	7.73×10^{-10}
3	10	1.76	0.72	2.12×10^{-9}	1.67	0.98	5.64×10^{-9}
4	30	1.33	2.00	6.02×10^{-8}	1.43	1.68	3.32×10^{-8}
5	50	0.76	3.72	6.77×10^{-7}	0.72	3.82	7.74×10^{-7}

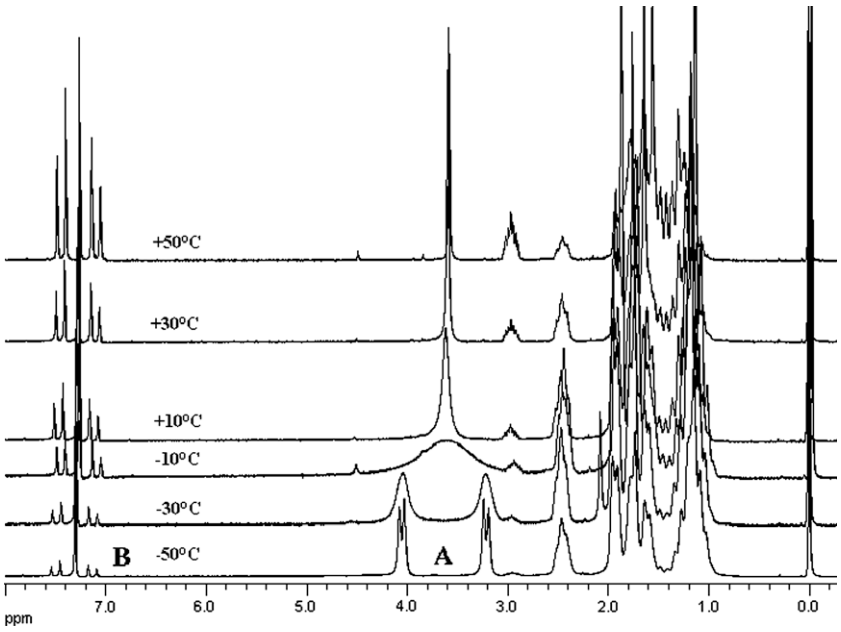


Fig. 4. ^{1}H NMR of **TCH** in $CDCl_3$ at various temperatures.

3.3. Behavior of 1,3,5-trialkyl-hexahydro-1,3,5-triazines in methanol

Dissolving **TCH** and **TH** in deuterated methanol results in an equilibrium mixture of **A**, **B** and **C**, which was confirmed on the basis of ^{1}H, ^{13}C, HSQC (Heteronuclear Single Quantum Coherence), HMBC (Heteronuclear Multiple Bond Correlation) and DEPT 135 (Distortionless Enhancement by Polarization Transfer) spectra of the **TCH** solution. The identification of the mixture components was made by analysis of signals corresponding to 2, 4, 6, 7, 8, 1′, 1″ and 1‴ H and C atoms (Fig. 5 and Table 4). The overlapping 2′–6′, 2″–6″ and 2‴–6‴ signals at 1–2 ppm in ^{1}H and 20–40 ppm in ^{13}C spectra were not essential for the consideration.

The presence of **A** in the mixture is evident from the C-2,4,6 methylene (negative-phased in DEPT 135) signal at 81.1 ppm correlating with 2,4,6-H at 4.17 ppm (HSQC) and 1′-H signal at 2.71 ppm (HMBC). The C-7 signal of the CH_2=N group in **B** (154.5 ppm) is correlated with 7-H$_\alpha$ at 7.45 ppm and 7-H$_\beta$ at 7.05 ppm (HSQC). 7-H$_\alpha$ correlates with C-1″ signal at 74.6 ppm (HMBC). The C-8 signal of compound **C** at 86.2 ppm (negatively-phased in DEPT 135) is correlated with 8-H at 4.32 ppm (HSQC) and 1‴-H signal at 2.78 ppm (HMBC). The 1‴-H signal, partly overlapped with 1′-H, is correlated with C-1‴ at 60.7 ppm (HSQC).

^{1}H NMR spectra of the mixture recorded at various temperatures confirmed the existence of a temperature-dependent **A**–**B**–**C** equilibrium (Scheme 2).

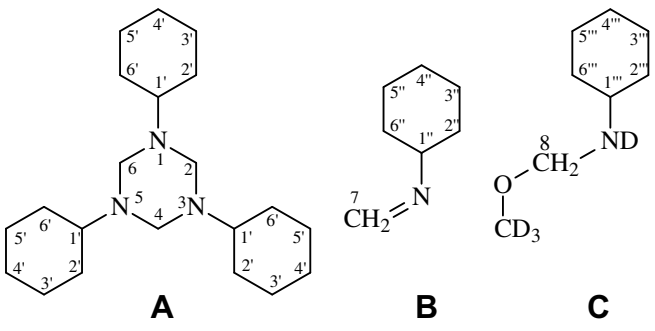


Fig. 5. Components of the mixture resulting from dissolving **TCH** (**A**) in CD_3OD.

The solution of **TCH** $[A_0] = 2 \times 10^{-4}$ mol dm^{-3} was prepared at room temperature and spectra taken at +50, +30, +10, −10 and −30 °C according to the description given for analogous investigation in $CDCl_3$ and $(CD_3)_2CO$. Due to the complexity of the system the calculations were limited to the molar fractions of **A**, **B** and **C.** The content of **A** in the mixture decreased with increasing temperature (Table 5 and Fig. 6).

The molar fraction of **C** is reasonably high at −30 °C, rises with increasing temperature reaching the highest recorded value of 0.59 at 30 °C, and then slightly decreases. The molar fraction of **A** decreases with increasing temperature from 0.54 at −30 °C to 0.24 at 50 °C. The molar fraction of **B** is lower than that of **A** and **C** at investigated conditions, but it rises with increasing temperature and at 50 °C is almost as high as the molar fraction of **A**.

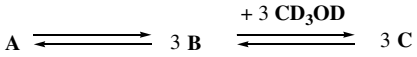
4. Conclusions

On the basis of the results it can be stated that the reversible depolymerization of 1,3,5-trialkyl-hexahydro-1,3,5-triazines (**A**) with formation of *N*-methylenealkylamines (**B**) takes place in inert solvents. The **A**/**B** ratio at equilibrium strongly depends on alkyl substituent. For linear ones, here represented by *n*-propyl, *n*-butyl and *n*-hexyl, the cyclic trimeric structures are highly stable and no signals of **B** could be observed in ^{1}H NMR spectra of the chloroform solutions at room temperature. For branched alkyl isomers such as *iso*-propyl, *tert*-butyl and cyclohexyl, considerable amount of form **B** could be detected in those solutions. The results of more detailed studies of the model compounds (**TH** and **TCH**) in a variety of inert solvents (chloroform, benzene, dimethylsulfoxide, acetone, acetonitrile) are in agreement with the above observation. Assuming that **B** are the active species in methylaminoalkylating reactions, the more branched isomers should react more easily. Further investigation of **A**–**B** equilibrium for **TCH** in acetone and chloroform revealed that lowering the initial concentration of the sample at room temperature results in **B** being the predominant form in both solutions (Table 2). With increasing temperature the amount of **B** in solution also rises (Table 3). 1,3,5-Trialkyl-hexahydro-1,3,5-triazines in methanol solution are also in equilibrium with **B**, however the latter readily transform into the corresponding *N*-(methoxymethyl)amines (**C**) in the reversible reaction with the

Table 4
^{1}H and ^{13}C NMR analytical signals and ^{1}H, ^{13}C correlations of the mixture components

	Atom C	δ (ppm)	Atom H	δ (ppm)	Multiple	^{1}H, ^{13}C HSQC	^{1}H, ^{13}C HMBC	DEPT 135
A	C-2,4,6	81.1	2,4,6-H	4.17	s	+	C-1′	↓
	C-1′	53.8	1′-H	2.71	m[a]	+	C-2,4,6	↑
	C-7	154.5	7-H$_\alpha$	7.45	d	+	C-1″	↓
B			7-H$_\beta$	7.05	d	+		
	C-1″	74.6	1″-H					↑
C	C-8	86.2	8-H	4.32	s	+	C-1‴	↓
	C-1‴	60.7	1‴-H	2.78	m[a]	+	C-8	↑

Abbreviations: s, singlet; d, doublet; m, multiplet. a, this signal overlapped with another signal; +, signal detected; ↑, positive-phased signal; ↓, negative-phased signal.



Scheme 2.

Table 5
Molar fractions of **A** (a), **B** (b) and **C** (c) depending on temperature

Entry	Temperature	a	b	c
1	−30	0.54	0.02	0.44
2	−10	0.49	0.05	0.46
3	10	0.40	0.11	0.49
4	30	0.28	0.13	0.59
5	50	0.24	0.19	0.57

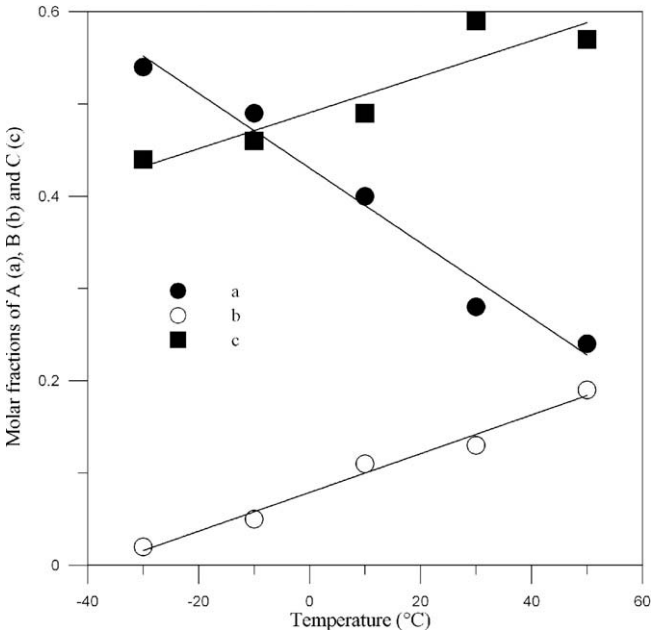


Fig. 6. Molar fractions of **A** (a), **B** (b) and **C** (c) depending on temperature.

solvent. The content of **C** in solution rises with increasing temperature and it becomes the main component of the mixture at room temperature.

Acknowledgements

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Appendix A. Supplementary data

The attribution of ^{1}H NMR and ^{13}C NMR signals of **A** and **B** in $CDCl_3$. ^{13}C NMR spectra of **TH** and **TCH** in $CDCl_3$. ^{13}C NMR, DEPT 135, HSQC, HMBC spectra of **TCH** in CD_3OD. ^{1}H NMR spectra of **TCH** in CD_3OD and $(CD_3)_2CO$ at −30, −10, +10, +30 and +50 °C. Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.molstruc.2008.05.019.

References

[1] E.M. Smolin, L. Rapoport, Heretocyclic Compounds: *s*-Triazines and Derivatives, Interscience Publishers, NY, 1959 (Chapter 9).
[2] M. Taguchi, N. Aikawa, G. Tsukamoto, Bull. Chem. Soc. Jpn. 61 (1988) 2431.
[3] A. Sucharda-Sobczyk, M. Wojciechowska, Chem. Stosowana 17 (1973) 359.
[4] J. Graymore, J. Chem. Soc. (1932) 1353.
[5] E.C. Wagner, J. Org. Chem. 19 (1954) 1862.
[6] D. Dickinson, J. Graymore, J. Chem. Soc. (1937) 1368.
[7] R.A.Y. Jones, A.R. Katritzky, M. Snarey, J. Chem. Soc. B (1970) 135.
[8] M.G. Ahmed, S.A. Ahmed, P.W. Hickmott, J. Chem. Soc., Perkin Trans. 1 (1980) 2383.
[9] J. Barluenga, A.M. Bayón, P. Campos, G. Asensio, E. Gonzales-Nuñes, Y. Molina, J. Chem. Soc., Perkin Trans. 1 (1988) 1631.
[10] E. Zangrando, G. Poggi, A.G. Giumanini, G. Verardo, J. Prakt. Chem. 329 (1987) 195.
[11] D.C. Frost, B. MacDonald, C.A. McDowell, N.P.C. Westwood, J. Electron Spectrosc. Relat. Phenom. 379 (1978) 222.
[12] J. Barluenga, A.M. Bayón, G. Asensio, J. Chem. Soc., Chem. Commun. (1984) 427.
[13] J.G. Guillemin, J.M. Denis, Tetrahedron 44 (1988) 4431.
[14] F.G. Riddell, J.M. Lehn, Chem. Commun. 12 (1966) 375.
[15] H.-J. Ha, W.K. Lee, Heterocycles 57 (2002) 1525.
[16] A.G. Giumanini, G. Verardo, E. Zangrando, L. Lassiani, J. Prakt. Chem. 329 (1987) 1087.
[17] H. Möhrle, K. Tröster, Arch. Pharm. 315 (1982) 619.
[18] G. Mlostoń, T. Gendek, Helv. Chim. Acta 81 (1998) 1585.
[19] H. Brunner, A. Winter, B. Nuber, J. Organomet. Chem. 558 (1998) 213.
[20] J. Wang, H. Leeman, R.A. Schoonheydt, Catal. Today 112 (2006) 188.
[21] R.D. Köhn, M. Haufe, S. Mihan, D. Lilge, Chem. Commun. (2000) 1927.
[22] R.D. Köhn, D. Smith, M.F. Mahon, M. Prinz, S. Mihan, G. Kociok-Köhn, J. Organomet. Chem. 68 (2003) 3200.
[23] R.T. Lewis, W.B. Motherwell, Tetrahedron 48 (1992) 1465.
[24] Y. Ohshiro, M. Komatsu, T. Agawa, Synthesis (1971) 89.
[25] A.G. Giumanini, G. Verardo, S. Cauci, J. Prakt. Chem. 329 (1987) 417.
[26] J.N. McKnight, E.S. Tillman, L.R. Sarry, Macromol. Rapid Commun. 27 (2006) 1578.
[27] H.-J. Ha, Ch.-J. Choi, Y.-G. Ahn, H. Yun, Y. Dong, W.K. Lee, J. Org. Chem. 65 (2000) 8384.
[28] K. Ikeda, T. Morimoto, M. Sekiya, Chem. Pharm. Bull. 28 (1980) 1178.
[29] H.-J. Ha, G.-S. Nam, K.P. Park, Synth. Commun. 21 (1991) 155.
[30] H.-J. Ha, K.-H. Kang, Y.-G. Ahn, S.-J. Oh, Heterocycles 45 (1997) 277.
[31] H.-J. Ha, Y.-S. Lee, Y.-G. Ahn, Heterocycles 45 (1997) 2357.
[32] [a] A. Adamczyk, Ph.D. Thesis, Warsaw University of Technology, 2007.; [b] K. Bujnowski, L. Synoradzki, A. Adamczyk-Woźniak, Arkivoc, accepted for publication.
[33] J.G. Jewett, J.J. Breeyear, J.H. Brown, C.H. Bushweller, J. Am. Chem. Soc. 122 (2000) 308.
[34] E. Müller, R. Kettler, M. Wiessler, Liebigs Ann. Chem. 8 (1984) 1468.